arXiv:quant-ph/9508024v1 24 Aug 1995

COLBY-94-02
IUHET 275

LONG-TERM EVOLUTION AND REVIVAL STRUCTURE OF RYDBERG WAVE PACKETS

Robert Bluhm[a] and V. Alan Kostelecký[b]

[a]*Physics Department*
Colby College
Waterville, ME 04901, U.S.A

[b]*Physics Department*
Indiana University
Bloomington, IN 47405, U.S.A.

It is known that, after formation, a Rydberg wave packet undergoes a series of collapses and revivals within a time period called the revival time, $t_{\rm rev}$, at the end of which it is close to its original shape. We study the behavior of Rydberg wave packets on time scales much greater than $t_{\rm rev}$. We show that after a few revival cycles the wave packet ceases to reform at multiples of the revival time. Instead, a new series of collapses and revivals commences, culminating after a time period $t_{\rm sr} \gg t_{\rm rev}$ with the formation of a wave packet that more closely resembles the initial packet than does the full revival at time $t_{\rm rev}$. Furthermore, at times that are rational fractions of $t_{\rm sr}$, the square of the autocorrelation function exhibits large peaks with periodicities that can be expressed as fractions of the revival time $t_{\rm rev}$. These periodicities indicate a new type of fractional revival occurring for times much greater than $t_{\rm rev}$. A theoretical explanation of these effects is outlined.

Published in Physics Letters A **200***, 308 (1995)*

An electron wave packet in a Rydberg atom provides a physical system in which to explore the interface between classical and quantum mechanics. When a Rydberg atom is excited by a short laser pulse, a state is created that has classical behavior for a limited time [1, 2]. Depending on the excitation scheme, either a radially localized wave packet in an eigenstate of angular momentum [3, 4, 5] or a packet localized in the angular coordinates [6, 7] is produced. Either a radial or a circular wave packet initially oscillates with the classical keplerian period $T_{\rm cl} = 2\pi\bar{n}^3$, where $\bar{n}$ is the central value of the principal quantum numbers excited in the packet. However, after a finite number of classical orbits quantum-interference phenomena appear and the wave packet collapses.

On a time scale large compared to the classical period $T_{\rm cl}$, a Rydberg wave packet passes through a sequence of fractional and full revivals [1, 2, 8, 9]. A full revival occurs at a time[1] $t_{\rm rev} = \frac{2\bar{n}}{3}T_{\rm cl}$, when a packet close to the original shape reappears. The fractional revivals occur earlier than this, at times that are rational fractions of the revival time $t_{\rm rev}$. They correspond to the formation of macroscopically distinct subsidiary wave packets that oscillate with periodicity that is a fraction of the classical orbital period $T_{\rm cl}$ [8]. Recent experiments have detected fractional revivals with periodicities as small as $T \approx \frac{1}{7}T_{\rm cl}$ [10].

In this letter, we examine the time evolution and revival structure of both radial and circular Rydberg wave packets on time scales much greater than the revival time $t_{\rm rev}$. A new system of full and fractional revivals is uncovered, with structure different from that of the usual fractional revivals.

In Ref. [1], long-term revivals at $t = \bar{n}^2 T_{\rm cl}$ and $t = \bar{n}^3 T_{\rm cl}$ were found. They were obtained by expanding the energy in a Taylor series in n through finite order and determining the times at which the time-dependent phase in the wave function is an integer multiple of 2π. Ref. [11] generalized this approach and found a hierarchy of recurrence times for full revivals of the wave packet for $t \gg t_{\rm rev}$.

Our approach here is different. We do not look for commensurability of the terms

[1] Note that $t_{\rm rev}$ is often defined as half the full revival time, since this is the time at which a full wave packet reforms for the first time, though with a phase difference relative to the classical motion. We adopt the notation of Ref. [8] for labeling the fractional revivals and use their definition of $t_{\rm rev}$.

in the phase with integer multiples of 2π. Rather, we show that at certain times greater than $t_{\rm rev}$ the wave function can be expanded in a set of subsidiary waves. This method is suitable for studying fractional revivals [8] as well as full revivals. It also reveals the presence of a long-term full revival occurring earlier than those found previously and not among the hierarchy of levels noted in Ref. [11].

A simple way of demonstrating the formation of the new fractional and full revivals is to construct the autocorrelation function of the time-evolved wave function with the initial packet. At the various fractional-revival times, the absolute square of the autocorrelation function exhibits periodicities reflecting the presence of underlying subsidiary wave packets. Furthermore, the ionization signal in a pump-probe experiment should display the same periodicities as the autocorrelation function. Thus, the primary features of our results relevant for experiments on Rydberg wave packets can be exhibited directly in a graph of the autocorrelation function as a function of time. For this reason, we first present plots of the autocorrelation function for times greater than $t_{\rm rev}$. We use these to indicate the structure of the new revivals. We then outline the theoretical explanation of the effects and suggest experiments to detect them.

Provided the eigenenergies are independent of the angular-momentum quantum numbers, it can be shown that the autocorrelation function for a given radial packet is the same as that for a related circular packet. For example, in the case of hydrogen the expression for the modulus squared of the autocorrelation function is

$$|A(t)|^2 = \Big| \sum_n \mid c_n \mid^2 e^{-iE_n t} \Big|^2 \tag{1}$$

for both types of wave packet. Here, $c_n = \langle \Psi(0) | \Psi_n \rangle$, and Ψ_n is an energy eigenstate with principal quantum number n.

We consider Rydberg wave packets of hydrogen for the case where the excitation spectrum is strongly peaked around a central value $\bar{n}$. For simplicity in the present work, we take a normalized gaussian distribution of width σ for the modulus squared of the coefficients, $|c_n|^2$. However, the results we obtain hold equally well for asymmetric excitation distributions, such as those occurring in a description in terms of radial squeezed states [12]. Our results also can be generalized to the case of nonin-

teger values of $\bar{n}$, including thereby the effects of quantum defects. This means, for example, that our results apply equally well to alkali-metal atoms. Experimentally, the value of σ is set by the exciting laser pulse. In what follows, the values chosen for σ correspond to a laser-pulse duration that is effective in displaying the new revival structure.

The time-dependent wave function for the Rydberg wave packet may be written as an expansion in terms of eigenstates of hydrogen

$$\Psi(\vec{r},t) = \sum_{k=-\infty}^{\infty} c_k \varphi_k(\vec{r}) \exp\left(-iE_n t\right) \quad , \tag{2}$$

where $k = n - \bar{n}$, $\bar{n}$ is assumed large, and $\varphi_k(\vec{r})$ is a hydrogenic wave function. Since we are taking a distribution strongly centered around $\bar{n}$, we may expand E_n around $\bar{n}$:

$$\Psi(\vec{r},t) = \sum_{k=-\infty}^{\infty} c_k \varphi_k(\vec{r}) \exp\left[-2\pi i \left(\frac{kt}{T_{\mathrm{cl}}} - \frac{k^2 t}{t_{\mathrm{rev}}} + \frac{k^3 t}{t_{\mathrm{sr}}} - \ldots + \ldots\right)\right] \quad . \tag{3}$$

In this equation, we have introduced three distinct time scales T_{cl}, t_{rev}, and t_{sr} that naturally appear in the first three orders in the expansion. The first two are the usual time scales relevant in the description of the conventional revival structure. The smallest time scale, T_{cl}, is the classical period for an electron in a keplerian orbit. If this term alone appeared in the expansion, the energies would be equally spaced and the wave packet would undergo simple harmonic motion without changing shape. The second term defines the revival time scale, t_{rev}, and is responsible for the collapse and fractional/full revivals of the wave packet [8].

The third-order term is included because in this paper we are interested in times much greater than the revival time. This term defines a new time scale, which we refer to as the superrevival time t_{sr}. In terms of the revival time, it is given by $t_{\mathrm{sr}} = \frac{3\bar{n}}{4} t_{\mathrm{rev}}$. For typical values of $\bar{n}$, t_{sr} is between one and two orders of magnitude greater than the revival time t_{rev}. However, this is still several orders of magnitude less than the lifetime of the excited Rydberg atom. It is therefore reasonable to examine the behavior of the wave packet and determine the effects of the third-order term on the revival structure up to times of order t_{sr}.

With the gaussian distribution for $|c_n|^2$ discussed above, we can evaluate the absolute square of the autocorrelation function $|A(t)|^2$ directly from Eq. (1) and plot

the result. We begin with an example involving a circular wave packet that serves to illustrate some key features of the new revival structure. Subsequently, we examine a radial wave packet with a lower value of $\bar{n}$ of a type that could be experimentally created and observed.

Consider the circular wave packet treated in Ref. [7], with $\bar{n} = 320$, $l = m = n-1$, and $\sigma = 2.5$, which displays a clear sequence of conventional fractional/full revivals on the revival timescale $t_{\rm rev} \simeq 1.06$ μsec. Figure 1 shows the absolute square of the autocorrelation function for a Rydberg wave packet with $\bar{n} = 320$ for a total time period large compared to the revival time. In this case, $t_{\rm sr} \simeq 255$ μsec, and we consider times up to just beyond $\frac{1}{6}t_{\rm sr}$. Figure 1a shows the behavior of the wave packet for approximately the first 9 μsec. The full revival appears at $t_{\rm rev} \simeq 1.06$ μsec, and fractional revivals are evident at fractions of this value. The largest revival appears at $t = \frac{1}{2}t_{\rm rev}$, where the wave function has reformed into a single wave packet, but at a time that is out of phase with a classically propagated distribution [7].

As can be seen from the figure, there are four or five full revival cycles, with peaks gradually decreasing in size and losing the canonical periodicity. However, after about 7 μsec a new structure with a noncanonical periodicity is visible in the autocorrelation function. Similar structures reappear later, at times near 14, 21, 28, and 42.5 μsec in Figures 1b, 1c, 1d, and 1e, respectively. The amplitudes and periodicities of these structures change with time. At $t \simeq 42.5$ μsec, a series of peaks appears with maximum amplitude greater than that of the full revival at $t = t_{\rm rev}$.

The features of this illustrative example can be predicted theoretically. We have proved that at times $t \approx \frac{1}{q}t_{\rm sr}$, where q must be an integer multiple of 3, the wave packet can be written as a sum of macroscopically distinct wave packets. The proofs are rather technical and are presented elsewhere [13]. Explicitly, we find that near times $t \approx \frac{1}{q}t_{\rm sr}$ we may write

$$\Psi(\vec{r}, t) = \sum_{s=0}^{l-1} b_s \psi_{\rm cl}(\vec{r}, t + \tfrac{s\alpha}{l} T_{\rm cl}) \quad , \tag{4}$$

where the macroscopically distinct wave packets are defined by

$$\psi_{\rm cl}(\vec{r}, t) = \sum_{k=-\infty}^{\infty} c_k \varphi_k(\vec{r}) \exp\left[-2\pi i \left(\tfrac{kt}{T_{\rm cl}}\right)\right] \tag{5}$$

and where their weights b_s in the sum (4) are given by

$$b_s = \frac{1}{l} \sum_{k'=0}^{l-1} \exp \left[2\pi i \left(\frac{\alpha s}{l} k' + \frac{3\bar{n}}{4q} k'^2 - \frac{1}{q} k'^3 \right) \right] \quad . \tag{6}$$

The quantities α and l are integer constants that depend on q and $\bar{n}$:

$$\alpha = \frac{2\bar{n}}{N} \quad , \qquad l = \begin{cases} q & \text{if } \; q/9 \neq 0 \pmod 1 \; , \\ q/3 & \text{if } \; q/9 = 0 \pmod 1 \; , \end{cases} \tag{7}$$

where N is an integer consisting of the product of all factors of $2\bar{n}$ that are also factors of l.

We have shown that the properties of the b_s coefficients are such that the autocorrelation function is periodic, with a period[2] $T \approx \frac{3}{q} t_{\rm rev}$. These periodicities are on a much greater time scale than the usual fractional revivals, which have periodicities that are fractions of $T_{\rm cl}$. Moreover, for special values of q the number of nonzero b_s coefficients is small and one coefficient dominates. This means that the wave function is mostly concentrated in one packet, which explains the peaks in the autocorrelation function. In special cases, only one nonzero b_s value exists. This first happens when $t \approx \frac{1}{6} t_{\rm sr}$, so that $q = 6$. It corresponds to the formation of a single packet, i.e., a full superrevival, with periodicity $T \approx \frac{1}{2} t_{\rm rev}$ in the autocorrelation function. For the fractional times $t \approx \frac{1}{q} t_{\rm sr}$ with $q > 6$, we find that more than one b_s coefficient is nonzero, corresponding to the formation of distinct subsidiary wave packets at times $t \gg t_{\rm rev}$. We refer to these as fractional superrevivals.

Since the b_s coefficients in (4) have moduli that are generally not equal, this indicates that the fractional superrevivals consist of an unequally weighted superposition of subsidiary wave packets, which is a feature that is not true for the usual fractional revivals. Moreover, after the formation of a fractional superrevival consisting of several subsidiary wave packets, it often happens that the subsidiary packets quickly evolve into a configuration where one of them is much greater than the others. The dominant wave packet in this case can again resemble the initial wave packet more closely than the full revival does at time $t_{\rm rev}$. In this way, large peaks can form in the autocorrelation function near the fractional superrevival times.

[2] The actual periodicity is $T = \frac{3}{q} t_{\rm rev} + \frac{u}{v} T_{\rm cl}$, where u and v are integers that depend on $\bar{n}$ and q. Since $T_{\rm cl} \ll t_{\rm rev}$, we write $T \approx \frac{3}{q} t_{\rm rev}$.

We demonstrate these results for the illustrative example with $\bar{n} = 320$. In this case, the theory predicts the first full superrevival appearing at $t \simeq 42.5$ μsec, with an autocorrelation-function periodicity $T \simeq 530$ nsec. This agrees with the structure seen in Figure 1e. For $q = 36$, 18, 12, and 9, fractional superrevivals are predicted to appear, since for these values of q more than one of the b_s coefficients is nonzero. The corresponding times at which structures should be seen are $t \simeq 7.08$, 14.2, 21.2, and 28.3 μsec, and they should have autocorrelation-function periodicities $T \approx \frac{1}{12} t_{\rm rev}$, $\frac{1}{6} t_{\rm rev}$, $\frac{1}{4} t_{\rm rev}$, and $\frac{1}{3} t_{\rm rev}$, respectively. These predictions agree with the structures seen in Figs. 1a – d.

To obtain further insight into the formation of a superrevival, we can examine the cross-sectional view of a circular wave packet at various times. Figure 2 shows a cross-sectional slice of the circular wave packet with $\bar{n} = 320$. We view the wave packets in the plane of the orbit and plot a circular slice $\Psi(\phi)$ at a fixed radius given by the expectation value $\langle r \rangle = \frac{1}{2} \bar{n}(2\bar{n} + 1)$. Figure 2a shows the initial wave packet at time $t = 0$. Figure 2b shows the wave packet at the canonical revival time $t \approx t_{\rm rev}$ when the autocorrelation function is a local maximum. As can be observed, the wave packet has indeed reformed and resembles the original wave, but it is asymmetric and several smaller subsidiary wave packets also appear. Figure 2c shows the full superrevival at $t \approx \frac{1}{6} t_{\rm sr}$, at which point only one b_s coefficient in Eq. (4) is nonzero. In comparing Figures 2b and 2c, the superrevival wave packet is seen to resemble more closely the initial wave packet of Figure 2a than does the canonical revival packet.

We chose $\bar{n} = 320$ as an illustrative example both for purposes of comparison with previous results for $t \leq t_{\rm rev}$ and because the full and fractional superrevivals are more evident for larger $\bar{n}$. However, similar effects can be observed for Rydberg wave packets with relatively small values of $\bar{n}$.

Figure 3 shows the square of the autocorrelation function for hydrogen with $\bar{n} = 48$ and $\sigma = 1.5$. As remarked above, the autocorrelation function for hydrogen is the same for both radial and circular wave packets. Thus, Figure 3 represents the autocorrelation function of a radial Rydberg wave packet that has been excited by a short laser pulse centered on the value $\bar{n} = 48$. In this case, the full revival is at $t \approx t_{\rm rev} \simeq 0.538$ nsec. For times greater than this, one can observe a fractional

superrevival at $t \approx \frac{1}{12}t_{\rm sr} \simeq 1.61$ nsec with autocorrelation periodicity $T \approx \frac{1}{4}t_{\rm rev}$ and a full superrevival at $t \approx \frac{1}{6}t_{\rm sr} \simeq 3.23$ nsec with autocorrelation periodicity $T \approx \frac{1}{2}t_{\rm rev}$. The size of the peak in the autocorrelation function shows that the superrevival resembles the initial wave packet more closely than does the revival wave packet at $t \approx t_{\rm rev}$.

We have proved that similar results hold when quantum defects are present in the energies. Therefore, we expect superrevivals to occur for wave packets in alkali-metal atoms as well as in hydrogen [14]. In alkali-metal atoms, the associated characteristic times depend on $\bar{n}^* = \bar{n} - \delta(l)$ instead of $\bar{n}$, where $\delta(l)$ is the quantum defect.

Figure 3 indicates it is likely an experiment can be performed to detect the full and fractional superrevivals discussed in this paper. One possibility is to use the pump-probe time-delayed photoionization method of detection for radial Rydberg wave packets excited in alkali-metal atoms with $\bar{n} \approx 45 - 50$. Single-photon absorption would produce a packet with localization purely in the radial coordinate and with the angular structure of a p state. The procedure is experimentally feasible, provided a delay line of 3 – 4 nsec is installed in the apparatus. In such an experiment, the fractional superrevival at $\frac{1}{12}t_{\rm sr}$, with periodicity $T \approx \frac{1}{4}t_{\rm rev}$, could be observed with a delay line of approximately 1.5 nsec. For even smaller values of $\bar{n}$, the required delay times can be reduced below 1 nsec. With $\bar{n} \simeq 36$, for example, the full/fractional superrevivals could be detected with delay lines used currently in experiments.

We enjoyed conversations with Charlie Conover and Duncan Tate. R.B. thanks Colby College for a Science Division Grant and the Theory Group at Indiana University for its hospitality.

1. J. Parker and C.R. Stroud, Phys. Rev. Lett. **56**, 716 (1986); Phys. Scr. **T12**, 70 (1986).
2. G. Alber, H. Ritsch, and P. Zoller, Phys. Rev. A **34**, 1058 (1986); G. Alber and P. Zoller, Phys. Rep. **199**, 231 (1991).
3. A. ten Wolde, L.D. Noordam, A. Lagendijk, and H.B. van Linden van den Heuvell, Phys. Rev. Lett. **61**, 2099 (1988).

4. J.A. Yeazell, M. Mallalieu, J. Parker, and C.R. Stroud, Phys. Rev. A **40**, 5040 (1989); J.A. Yeazell, M. Mallalieu, and C.R. Stroud, Phys. Rev. Lett. **64**, 2007 (1990); J.A. Yeazell and C.R. Stroud, Phys. Rev. A **43**, 5153 (1991).
5. D.R. Meacher, P.E. Meyler, I.G. Hughes, and P. Ewart, J. Phys. B **24**, L63 (1991).
6. J.A. Yeazell and C.R. Stroud, Phys. Rev. Lett. **61**, 2099 (1988).
7. Z. Dačić Gaeta and C.R. Stroud, Phys. Rev. A **42**, 6308 (1990); Z. Dačić Gaeta, M. Noel, and C.R. Stroud, unpublished.
8. I.Sh. Averbukh and N.F. Perelman, Phys. Lett. **139A**, 449 (1989).
9. M. Nauenberg, J. Phys. B **23**, L385 (1990).
10. J. Wals, H.H. Fielding, J.F. Christian, L.C. Snoek, W.J. van der Zande, and H.B. van Linden van den Heuvell, Phys. Rev. Lett. **72**, 3783 (1994).
11. A. Peres, Phys. Rev. A **47**, 5196 (1993).
12. R. Bluhm and V.A. Kostelecký, Phys. Rev. A **48**, R4047 (1993) (quant-ph/9508019); Phys. Rev. A **49**, 4628 (1994) (quant-ph/9508020).
13. R. Bluhm and V.A. Kostelecký, Phys. Rev. A **51**, 4767 (1995) (quant-ph/9506009).
14. R. Bluhm and V.A. Kostelecký, Phys. Rev. A **50**, R4445 (1994) (hep-ph/9501372).

Fig. 1: The absolute square of the autocorrelation function for a Rydberg wave packet with $\bar{n} = 320$ and $\sigma = 2.5$ is plotted as a function of time in microseconds. The value $\sigma = 2.5$ for a wave packet with $\bar{n} = 320$ corresponds to excitation with a 160 picosecond laser pulse. (a) $0 \leq t \leq 9$ μsec, (b) $9 \leq t \leq 18$ μsec, (c) $18 \leq t \leq 27$ μsec, (d) $27 \leq t \leq 36$ μsec, (e) $36 \leq t \leq 45$ μsec.

Fig. 2: Unnormalized circular wave packets with $\bar{n} = 320$ and $\sigma = 2.5$. Cross-sectional slices of the wave packet in the plane of the orbit and for $r = \langle r \rangle = \frac{1}{2}\bar{n}(2\bar{n}+1)$ are plotted as a function of the azimuthal angle ϕ in radians. (a) $t = 0$, (b) $t \approx t_{\rm rev}$, (c) $t \approx \frac{1}{6}t_{\rm sr}$.

Fig. 3: The absolute square of the autocorrelation function for a Rydberg wave packet with $\bar{n} = 48$ and $\sigma = 1.5$ is plotted as a function of time in nanoseconds. The value $\sigma = 1.5$ for a wave packet with $\bar{n} = 48$ corresponds to excitation with a 900 femtosecond laser pulse.

This figure "fig1-1.png" is available in "png" format from:

http://arXiv.org/ps/quant-ph/9508024v1

This figure "fig2-1.png" is available in "png" format from:

http://arXiv.org/ps/quant-ph/9508024v1

This figure "fig1-2.png" is available in "png" format from:

http://arXiv.org/ps/quant-ph/9508024v1

This figure "fig2-2.png" is available in "png" format from:

http://arXiv.org/ps/quant-ph/9508024v1

This figure "fig1-3.png" is available in "png" format from:

http://arXiv.org/ps/quant-ph/9508024v1

This figure "fig2-3.png" is available in "png" format from:

http://arXiv.org/ps/quant-ph/9508024v1

This figure "fig1-4.png" is available in "png" format from:

http://arXiv.org/ps/quant-ph/9508024v1

This figure "fig2-4.png" is available in "png" format from:

http://arXiv.org/ps/quant-ph/9508024v1